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Endocrine Disruptors and Asthma-Associated Chemicals in Consumer Products

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Running Title

Chemicals in consumer products

Keywords

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Abbreviations

AHTN	acetyl methyl tetramethyl tetralin
APEO	alkylphenol ethoxylate
BBP	benzylbutyl phthalate
BP-1	benzophenone-1
BP-3	benzophenone-3
BPA	bisphenol A
CDC	Centers for Disease Control and Prevention
cVMs	cyclic volatile methylsiloxanes
D3	hexamethylcyclotrisiloxane
D4	octamethylcyclotetrasiloxane
D5	decamethylcyclopentasiloxane
D6	dodecamethylcyclohexylsiloxane
DBP	di-n-butyl phthalate
DCP	di-cyclohexyl phthalate
DEA	diethanolamine
DEHP	bis-2-ethylhexyl phthalate
DEP	diethyl phthalate
DINP	di-isononyl phthalate
DL	detection limit
DPP	di-n-propyl phthalate
EDCs	endocrine disrupting compounds
FDA	Food and Drug Administration
FIFRA	Federal Insecticide, Fungicide and Rodenticide Act
HHCB	hexahydrohexamethyl cyclopentabenzopyran
MEA	monoethanolamine
NHANES	National Health and Nutrition Examination Survey
NP	4- <i>t</i> -nonylphenol
NP1EO	nonylphenol monoethoxylate
NP2EO	nonylphenol diethoxylate
PBDE	polybrominated diphenyl ether
PCB	polychlorinated biphenyl
QA/QC	quality assurance / quality control
TEA	triethanolamine
UV	Ultraviolet
VOCs	volatile organic compounds

Abstract

BACKGROUND: Laboratory and human studies raise concerns about endocrine disruption and asthma from exposure to chemicals in consumer products. Limited labeling or testing information is available to evaluate products as exposure sources.

OBJECTIVES: We analytically quantified endocrine disruptors and asthma-related chemicals in a range of cosmetics, personal care products, cleaners, sunscreens, and vinyl products. We evaluated whether labels can be used to select products without these chemicals.

METHODS: We selected 213 commercial products representing 50 product types. We tested 42 composited samples of high market-share products. We also tested 43 alternative products identified using criteria expected to minimize target compounds. Analytes included parabens, phthalates, bisphenol A, triclosan, ethanolamines, alkylphenols, fragrances, glycol ethers, cyclosiloxanes and UV filters.

RESULTS: We detected 55 compounds, indicating a wide range of exposures from common products. Vinyl products contained >10% DEHP and could be an important source of DEHP in homes. In other products, the highest concentrations and numbers of detects were in the fragranced products perfume, air fresheners, and dryer sheets, and in sunscreens. Some products that did not contain the well-known EDC phthalates contained other less-studied phthalates (also EDCs), suggesting a substitution. Many detected chemicals were not listed on labels.

CONCLUSIONS: Common products contain complex mixtures of EDCs and asthma-related compounds. Toxicological studies of these mixtures are needed to understand their biological activity. For epidemiology, findings raise cautions about potential confounding from co-occurring chemicals and misclassification due to variability in product composition. It appears that consumers can avoid some target chemicals—synthetic fragrances, BPA, and regulated active ingredients—using purchasing criteria. More complete labeling would enable consumers to avoid the rest.

Introduction

Consumer product chemicals are ubiquitously detected, sometimes at high concentrations, in human tissues (CDC 2009) and household air and dust. (Rudel et al. 2003; Rudel and Perovich 2009; Rudel et al. 2010; Weschler 2009). Studies of pesticides, polychlorinated biphenyls (PCBs), polybrominated diphenyl ether (PBDE) flame retardants, and volatile organic compounds (VOCs) in homes provide some information about sources, exposure pathways, and exposure reduction options (Dodson et al. 2008; Lorber 2008; Rudel et al. 2008; Zota et al. 2008). However, for many common commercial chemicals, information about the contribution of specific consumer products to exposure is limited. In particular, little information is available about exposures from personal care and cleaning products.

Many of these products may be sources of chemicals that have a diverse spectrum of health effects, including endocrine disruption and associations with asthma. Endocrine disrupting compounds (EDCs) are chemicals that can alter hormonal signaling with potential effects on developing reproductive and nervous systems, metabolism, and cancer (Colborn et al. 1993). Some phthalates inhibit testosterone synthesis (Howdeshell et al. 2008), and antimicrobials like triclosan suppress thyroid hormone (Paul et al. 2010) and are estrogenic (Stoker et al. 2010) in mammalian models. Some parabens, alkylphenols, cyclosiloxanes, UV filters, and synthetic musk fragrance compounds are weakly estrogenic in a variety of experimental models (Bitsch et al. 2002; Bonefeld-Jorgensen et al. 2007; Quinn et al. 2007; Routledge et al. 1998; Schlumpf et al. 2004; Schreurs et al. 2005). Factors related to home environments are associated with asthma, although there has been limited study of the role of chemical contaminants (Douwes and Pearce 2002). Fragrances have been shown to exacerbate asthma (Kumar et al. 1995). The phthalate bis-2-ethylhexyl phthalate (DEHP) in dust was

associated with asthma and wheezing in children (Bornehag et al. 2004) and several phthalates show an adjuvant effect in animal studies (Bornehag and Nanberg 2010). The sum of propylene glycol and glycol ethers was associated with increased asthma prevalence in pre-school children (Choi et al. 2010). The ethanolamines monoethanolamine and diethanolamine are occupational asthmagens (Association of Occupational and Environmental Clinics 2010).

Previous research suggests that consumer products are a source of these compounds in homes. We found a wide range of phthalates, alkylphenols, parabens, flame retardants, PCBs, and current-use and banned pesticides in air and dust samples from homes, with 13-28 compounds in air and 6-42 in dust (Rudel et al. 2003). Analysis of paired indoor and outdoor air samples in California demonstrated indoor concentrations were considerably higher than outdoors for many compounds, indicating constant indoor sources (Brody et al. 2009; Rudel et al. 2010).

Efforts to identify the contribution of specific products to home environments or personal exposure are hindered by limited and inconsistent disclosure of chemical ingredients in consumer products. Regulations require only limited labeling. For example, sunscreens, antiperspirant deodorants, and anti-bacterial hand soaps are regulated as over-the-counter drugs by the US Food and Drug Administration (FDA) and “active” ingredients must be labeled (Food and Drug Administration 1938, 1967). For cosmetics, the FDA requires listing of ingredients in order of predominance, except chemical constituents of fragrances and “incidental ingredients” do not need to be listed (Food and Drug Administration 1938, 1967). For cleaning products, ingredient labeling is required only for compounds, such as antimicrobials, regulated by US EPA under the Federal Insecticide, Fungicide and Rodenticide Act (FIFRA) (Environmental Protection Agency 1972). The labeling terms “natural,” “non-toxic,” and “green” are unregulated and do not

provide standardized ingredient information. Indeed, a recent study found that the VOC composition of “green” labeled fragranced products was not significantly different from other fragranced products with regard to number of hazardous chemicals as defined under U.S. federal laws (Steinemann et al. 2010).

Gaps in ingredient information are problematic from multiple perspectives. Regulators rely on ingredient concentrations in products for exposure modeling. Consumers also want ingredient information to make precautionary choices consistent with personal values, and although environmental health organizations have developed rating systems to advise consumers, these are limited to information on labels (Environmental Working Group 2011; GoodGuide 2012). In addition, researchers need ingredient information to interpret health studies and test exposure reduction strategies. In an effort to fill this gap, we asked 34 manufacturers in 2007 to tell us whether specific personal care and cleaning products contained EDCs from a list we provided, but many were unwilling (Dunagan et al. 2011).

In order to develop information about exposure sources, we characterized the concentrations of 66 chemicals in 42 types of household products, focusing on cleaners and personal care products. We also aimed to identify the predominant exposure sources to target for product substitution in an intervention study. Such intervention designs are powerful approaches to exposure assessment and have been used to estimate bisphenol A (BPA) and phthalate exposure via food packaging (Rudel et al. 2011), and pesticide exposure from food (Lu et al. 2006). To identify substitute products to use in an intervention study, we measured concentrations of the same 66 chemicals in 43 “alternative” product samples, selected because their labels indicated that they might be free of the chemicals of concern. Thus, results also

provide insight into the usefulness of product labeling for consumers seeking to reduce exposures.

Methods

We selected the 66 organic chemicals for inclusion in the study based on evidence of endocrine disruption or asthma exacerbation, expected presence in consumer products, and compatibility with analytical methods developed in our household exposure studies (Rudel et al. 2003; Rudel et al. 2010). We tested 85 samples representing 213 products in two rounds of chemical analysis. Table S1 lists the chemical groups, their typical uses, and the evidence of endocrine disruption or asthma exacerbation.

Product Selection

We first identified the types of products likely to contain compounds of interest. Product types included personal care products (e.g., lotion, hair products, and toothpaste), cleaners (e.g., laundry detergent and all-purpose cleaner), and other household goods, and are listed in Figures 1-2. Then we identified several “conventional” products and one “alternative” for each product type. Criteria for alternative products are shown in Table 1. A product was classified as alternative if the label indicated that excluded chemicals were absent, or the label listed ingredients and did not list excluded terms. Many of the products that met our criteria for alternative products were marked as “green.” We also included as alternative products 6 items often used in recipes for home-made cleaners such as bleach and vinegar. Products that did not meet the “alternative” criteria were classified as conventional. In selecting conventional products, we aimed for products that are widely used in order to better represent typical exposures. We lacked comprehensive information from which to select products, so we

identified leading companies for the product sector (e.g., hair care) based on market share, and selected candidate products from several leading companies. When possible we also included a generic store-brand product. Final product selections were made informally, based on availability and shelf space. We purchased most alternative products at a nation-wide store specializing in natural products, so products met the store's selection criteria, which favored non-petroleum-based, and especially plant-based, ingredients. We purchased the majority of conventional products at major grocery and pharmacy chain stores. We purchased most products in fall 2007. We added products for a second round of chemical analysis approximately one year later.

Sampling Design and Compositing

We analyzed 42 analytical samples composited from 170 conventional products and 43 samples of individual alternative products.

Because we aimed to cost-effectively evaluate typical exposures from conventional products, we composited 170 conventional products into a single sample for each product type (42 analytical samples). We combined equal masses of 1 to 7 products within a product type and analyzed the mixture as a single sample. The advantage of compositing is that samples may provide more generalizable exposure information. However, composited samples are more limited in that they (a) will not reveal an unusually high concentration in a single product if that product is mixed with others having lower concentrations; (b) will not reveal a concentration just above the detection limit in a single product if that product is mixed with others having concentrations below detection; and (c) may show a higher detection frequency for chemicals well within the detectable range.

For alternative products, our goal was to identify specific products that were free of the chemicals of concern, so the products could be used in an intervention study. Thus, we analyzed

just one alternative product per product type (43 analytical samples, one for each of 43 individual products). Therefore, reported detection frequencies and concentrations for conventional and alternative product types are not directly comparable. To provide some information about variability in products within a category, we tested individual samples of five alternative sunscreens, and calculated an average for the product type “alternative sunscreen”.

Chemical Analysis

We analyzed samples in two rounds: 50 compounds in the first round and those 50 plus 16 other compounds in the second round. Products were composited as described, surrogate recovery standards added, and then samples were extracted with dichloromethane:methanol, passed through a weak anion exchange cartridge, spiked with internal standard, and analyzed by GC/MS in the full scan mode. A separate aliquot was derivitized and analyzed for phenolic compounds.

For each compound, the method reporting limit (MRL) was defined as the maximum of the analytical detection limit and the 90th percentile of the blank concentrations within each analytical round. The reporting limit was 1 µg/g for chemicals in products, and was reported above 1 µg/g if there were detectable concentrations in the blank samples (1 chemical in analytical round 1 and 12 in analytical round 2).

Extensive Quality Assurance/Quality Control (QA/QC) samples were included. Detects in blanks were infrequent and elevated MRLs were 5 µg/g or less except for the cyclosiloxane decamethylcyclopentasiloxane (D5; the only compound detected in >75% of blanks; results were blank corrected by subtracting the median blank value from the reported value). Precision was assessed with 13 duplicate samples (relative percent difference generally <50%), and accuracy

by determining spike recovery for all target compounds in 6 different matrices (median recoveries across products were generally within 50-150%) and by calculating recoveries of surrogates in all samples (median percent recoveries were within the 50-150% acceptance range for all surrogates over both analytical rounds). Additional details regarding chemical analysis and QA/QC measures can be found in Supplemental Material section Analytical Quality Assurance/Quality Control (QA/QC) Methods and Results beginning on page S-9.

Data Analysis

Our analysis of this large dataset is visual and exploratory. In Figures 1 and 2, we graphed product type against compounds detected using a “heat map” approach for conventional and alternative products, respectively. Only values above the MRL or 1 µg/g are presented. In Supplemental Material, Figure S1, we graphed results for sunscreens in a similar format. The graph presents results for a composited sample of conventional sunscreens, the calculated composite obtained by averaging results for five alternative sunscreens, and individual results for the five alternative sunscreens.

To identify chemicals that tend to co-occur because they are used together in a product, we estimated correlations for chemicals simultaneously detected within a product type (e.g., laundry detergent, lipstick). We calculated Kendall’s *tau* adjusted for censored data and with *p*-values obtained from 10,000 bootstrap replications (Newton and Rudel 2007). Note that the magnitude of the Kendall’s *tau* coefficients tends to be smaller than the more familiar Spearman’s correlation coefficients. We limited this analysis to chemicals detected in >3 analytical samples, and conducted the analysis separately for conventional and alternative products.

Results and Discussion

We tested 213 conventional and alternative consumer products in 85 analytical samples for up to 66 compounds of interest. We detected 55 compounds: 50 chemicals in 42 conventional samples representing 170 products, and 41 in 43 alternative samples representing 39 product types.

The most frequently detected compounds in conventional products were two natural fragrance compounds (may be derived from plant materials-further defined below in Fragrances section), two synthetic fragrance compounds, and diethyl phthalate (DEP) (Figure 1). The most frequently detected compounds in alternative products, including the calculated composite sunscreen, were two natural fragrance compounds, an alkylphenol, methyl paraben and DEP (Figure 2).

We detected 11 compounds at concentrations greater than 1% (10,000 µg/g) and 26 above 0.1%. DEHP was detected at 28% and 14% by weight in the vinyl shower curtain composite and vinyl pillow protector, respectively. The glycol ether 2-butoxyethanol was detected at nearly 5% in the carpet cleaner. The sunscreen samples had 2.5-6.2% of the ultra-violet (UV) filters octinoxate and benzophenone-3 (BP-3). The fragrance/perfume composite contained almost 3% HHCB, a synthetic fragrance chemical, and 1.4% DEP. The car air freshener contained the fragrance chemicals isobornyl acetate and limonene (a natural fragrance compound) at approximately 2% each. The alternative shampoo sample had 2.4% diethanolamine.

We describe additional findings below by chemical class. We then present results from the analysis of mixtures and finally discuss product variability using sunscreen as an example.

Parabens

Parabens are added to many consumer products, pharmaceuticals, and foods as preservatives and anti-microbial agents (Soni et al. 2001). Previous studies found parabens, particularly methyl paraben, in most cosmetic samples (Rastogi et al. 1995; Shen et al. 2007). Methyl and propyl paraben were detected in over 96% of urine samples in a recent study of 100 demographically diverse adults (Ye et al. 2006). Parabens are weakly estrogenic *in vitro* and butyl paraben was reported to affect reproductive tract development in rats at 100 mg/kg doses (Kang et al. 2002).

We detected parabens in personal care products but not cleaners. Methyl paraben was detected most frequently and at the highest concentrations; ethyl- and butyl paraben were only found if methyl paraben was also detected. The highest concentration was in an alternative sunscreen (methyl paraben; 1600 µg/g). Of the 11 conventional samples with detectable parabens, 10 included products with “paraben” on the label. With the exception of shaving cream, products were not considered alternative if parabens were listed as an ingredient. Nevertheless, in alternative products, we detected parabens in 7 products, including 3 sunscreens, that did not list parabens on the label.

Phthalates

Phthalates are used as plastic additives, solvents in cosmetics and perfumes, and as an inert ingredient in pesticides. Higher molecular weight phthalates (e.g., DEHP) are typically used in plastics (10-60% by weight) and readily migrate out of products (Rakkestad et al. 2007). Lower molecular weight phthalates (e.g. di-n-butyl phthalate (DBP), DEP) are typically used as solvents in personal care products and in lacquers, varnishings, and coatings (Meeker et al. 2009b). A number of different phthalates have been reported in cosmetic and other personal care

products, sometimes at concentrations above 1% (Hubinger and Havery 2006; Koniecki et al. 2011; Shen et al. 2007). Near universal detection of phthalates in urine samples shows widespread exposure (CDC 2009; Heudorf et al. 2007). Phthalates have been associated in humans with adverse reproductive system outcomes, including reduced semen quality and altered male genital development, as well as respiratory symptoms (Bornehag et al. 2004; Engel et al. 2010; Hauser and Calafat 2005; Hauser et al. 2006; Kimber and Dearmna 2010; Meeker et al. 2009a; Meeker et al. 2009b; Mendiola et al. 2011; Swan et al. 2005; Swan 2008). Many phthalates are identified as anti-androgenic EDCs in mammalian models, while DEP is not generally characterized as an endocrine active compound (Hannas et al. 2011; Heindel et al. 1989; Howdeshell et al. 2008). Among the EDCs in this study, phthalates are the only chemical group for which there is supporting evidence of health effects from human studies.

We analyzed samples for 12 phthalates. DEP, a common solvent for fragrance (Hubinger and Havery 2006), was detected most frequently; the highest DEP concentrations were in fragrance/perfume (14,000 µg/g) and car air freshener (8,000 µg/g). Vinyl products had the highest concentrations of any phthalate, with DEHP at 28% in the shower curtains and 14% in pillow protector. DBP and benzylbutyl phthalate (BBP) were detected in conventional pillow protector, dryer sheet, polish/wax, car interior cleaner, and in alternative tub/tile cleaner, bar soap, shaving cream, and lipstick. Three phthalates were found only in alternative products: di-cyclohexyl phthalate (DCP), di-isononyl phthalate (DINP), and di-n-propyl phthalate (DPP). These compounds may have been introduced as substitutes for the better-known anti-androgenic phthalates (DBP, BBP, DEHP), even though they are also EDCs, with similar (DCP) or lesser (DINP, DPP) potency (Boberg et al. 2011; Hannas et al. 2011; Heindel et al. 1989; Saillenfait et al. 2009). The alternative shaving cream contained 5 different phthalates, illustrating the

potential for simultaneous exposures to multiple phthalates, which act cumulatively on endocrine targets (NRC 2008). No products we tested had “phthalate” on the label, including personal care products where intentional ingredients must be labeled. However, the conventional nail polish sample with measurable DEHP contained a product labeled with “phthalic anhydride copolymer.”

Bisphenol A

BPA is used in a variety of consumer products containing epoxy resins, polyester-styrene, and polycarbonate plastics. It can be an ingredient in vinyl and in dental sealants, protective coatings, flame retardants, and adhesives (Meeker et al. 2009b). Biomonitoring studies indicate that exposure is widespread; BPA was detected in over 93% of urine samples in the National Health and Nutrition Examination Survey (NHANES) (Calafat et al. 2008c). A wide body of laboratory evidence shows BPA-induced endocrine disruption in a number of organ systems (FAO/WHO 2010).

We detected BPA in 15 conventional samples including the vinyl shower curtain and pillow protector, dish and laundry detergent, tub and tile cleaner, soaps, lotions, shampoo, conditioner, shaving cream, nail polish, and sunscreen. Concentrations were under 100 µg/g, with most less than 10 µg/g. BPA was not detected in alternative samples except sunscreen, so selecting alternative products according to our criteria appears to avoid BPA. No labels listed BPA.

Antimicrobials

We analyzed four antimicrobials: ortho-phenylphenol, triclocarban, triclosan, and 1,4-dichlorobenzene. Both triclocarban and triclosan are broad-spectrum agents commonly added to

personal care products, such as toothpastes and soaps; detergents; toys; plastics; and textiles (Adolfsson-Erici et al. 2002; Calafat et al. 2008b; Perencevich et al. 2001). A national survey detected triclosan and triclocarban in 76% of liquid soaps and 29% of bar soaps (Perencevich et al. 2001) and triclosan was detected in 76% of NHANES urine samples (Calafat et al. 2008b). Triclosan has been shown to disrupt thyroid homeostasis in mammalian models (Paul et al. 2010; Rodriguez and Sanchez 2010) and current human exposure levels are in the range of those predicted to have this activity based on laboratory tests (Rotroff et al. 2010). Triclocarban has been shown to amplify endogenous androgen response in mammalian models (Chen et al. 2008). Personal care product labels must list antimicrobial concentrations (FDA 2009).

We detected triclocarban and triclosan, but not the other two antimicrobials. When they were detected at higher concentrations, product labels listed them as active ingredients, consistent with FDA labeling requirements. For example, the conventional bar soap sample contained triclocarban at 1520 µg/g and one of the 4 soaps in the composite was labeled “antibacterial” with triclocarban (0.6%). Concentrations of triclosan in conventional hand soap and toothpaste were slightly lower than predicted from labeling of active ingredient concentrations. Some products (conventional facial cleanser and lipstick) listed triclosan in the ingredient list, but not as an active ingredient, but we did not detect it in those composite samples. Also, we found relatively low levels (6 µg/g) of triclosan in conventional dish liquid composite although it was not listed on the product labels. We did not detect these compounds in any of the alternative products.

Ethanolamines

Monoethanolamine (MEA) is used in cleaners and degreasers, detergents, soaps, cosmetics, hair dyes, and as an emulsifier in lotions and creams; and diethanolamine (DEA) is

used as an emulsifier in shampoos, cleaners, detergents, polishes, and auto products (National Library of Medicine 2010b). Exposure studies are limited. MEA and DEA have been associated with occupational asthma (Association of Occupational and Environmental Clinics 2010; Kamijo et al. 2009; Makela et al. 2011; Piipari et al. 1998; Savonius et al. 1994). The European Commission prohibits DEA in cosmetics and restricts products with MEA to less than 0.5% amine content because of concerns about formation of carcinogenic nitrosamines (European Commission 2011).

We detected MEA in conventional glass cleaner and laundry detergent ($>1000 \mu\text{g/g}$) and alternative surface cleaner, glass cleaner, and shampoo ($<400 \mu\text{g/g}$). The highest DEA concentration was in the alternative shampoo at $24,000 \mu\text{g/g}$, or 2.4% by weight. DEA was detected in the composite sample of conventional dryer sheets ($840 \mu\text{g/g}$) and 4 other conventional and alternative cleaning and personal care product samples ($<150 \mu\text{g/g}$): conventional face lotion and alternative tube and tile cleaner, mascara and shaving cream.

No labels listed MEA or DEA. Several product labels included the derivatives “cocamide MEA,” “lauramide DEA,” or “cocamide DEA,” but MEA and DEA were not detected in samples of these products. We detected DEA in mascara labeled with triethanolamine (TEA), but MEA and DEA were not detected in other TEA-labeled products. Commercial mixtures of TEA may contain small amounts of DEA and MEA (WHO 2000).

Alkylphenols

Alkylphenol polyethoxylates (APEOs), including nonylphenol and octylphenol ethoxylates, are used as surfactants in consumer products such as detergents, disinfectants, and surface cleaners, and as “inert” ingredients in pesticides. Mixtures with ethoxylate chains of

varying lengths are typical and can degrade to nonylphenol and octylphenol, both identified as weakly estrogenic (Jie et al. 2010). Nonylphenol also originates from vinyl and other plastics containing tris(nonylphenol) phosphite and may have other uses, including in epoxy resins. The branched chain para-substituted nonylphenol and octylphenol are the commercially prevalent compounds (European Commission 2002; Rudel and Perovich 2009; Rudel et al. 2010).

We measured 4-*t*-octylphenol and 4-*t*-nonylphenol (NP) and their mono and diethoxylates and detected them in about half of the samples, including plastics, cleaners and personal care products. Concentrations were low (<20 µg/g), except in the conventional car cleaner and vinyl shower curtain.

Product labels did not list alkylphenols. Of the 37 alkylphenol-containing samples, 7 included products labeled with “surfactants” of any type (e.g. ionic, non-ionic). Ten samples contained at least one product labeled with “surfactants” but alkylphenols were not detected. Since many products containing APEOs did not list surfactants on labels, a significant fraction of the products may contain NP from other uses, such as plasticizer, or are simply unlabeled. Thus it appears that exposure to alkylphenols cannot be avoided by label reading.

Fragrances

Fragrances are added to products to achieve a desired scent or mask other scents in the product. Over 3,000 fragrance ingredients have been reported and a fragranced product may contain 50-300 different chemicals (Bickers et al. 2003). Exact formulations are typically protected from disclosure (Bridges 2002; International Fragrance Association 2010). Fragrances can be either synthetic or natural/plant-based; however some natural fragrance chemicals can be artificially synthesized, and these may or may not reflect the natural stereoisomer composition

(Ravid et al. 2009) and may have different health effects (Smith 2009). In this study we classified fragrance chemicals as natural if they are readily available from plant materials and synthetic if they are most commonly synthesized, based on information in the Hazardous Substances Data Bank (National Library of Medicine 2010a), however we did not independently verify that the natural fragrance chemicals were not synthesized. Synthetic fragrance compounds, which include polycyclic and nitro musks, have been found in many personal care and household products and at concentrations up to several thousand micrograms per gram (Reiner and Kannan 2006; Zhang et al. 2008). Synthetic and natural fragrance compounds have been reported in cleaning products (Rastogi et al. 2001), cosmetics, and perfumes, and it should be noted that in a 1996 study a high proportion of perfumes and cosmetics labeled as containing natural ingredients in fact contained synthetic fragrances (Rastogi et al. 1996). Fragrances, particularly terpenes such as limonene, are associated with secondary chemical reactions in indoor air, and can contribute to the production of formaldehyde, glycol ethers, ultrafine particles, and secondary organic aerosols (Nazaroff and Weschler 2004; Singer et al. 2006). Exposure to fragrances has been associated with a range of health effects, including allergic contact dermatitis, asthma and asthmatic exacerbations, headaches, and mucosal symptoms (Heydorn et al. 2003; Kumar et al. 1995; Steinemann 2009). Synthetic musks have been shown to have estrogenic effects (Bitsch et al. 2002; Schreurs et al. 2005; Seinen et al. 1999; van der Burg et al. 2008).

We frequently detected synthetic and natural fragrance chemicals in conventional samples. In our alternative products selected to allow only plant-based fragrance, synthetic fragrance chemicals were detected only in the facial cleanser, floor cleaner and one sunscreen at less than 100 µg/g. Bucinal, HHCB and methyl ionone were the most frequently detected

synthetic fragrance chemicals in conventional product samples. Concentrations of these and AHTN, isobornyl acetate, and phenethyl alcohol included detects above 1000 µg/g in fragrance/perfume, car and home air fresheners, and dryer sheets. Natural fragrance chemicals were common in conventional and alternative products. Most common natural fragrance chemicals included the terpenes limonene, hexyl cinnamal, and linalool. Concentrations of fragrance compounds were generally higher in conventional (21 instances > 1000 µg/g) than alternative products (2 instances > 1000 µg/g), reflecting that product types used specifically to create scent (e.g. air freshener, perfume) were categorized as conventional and can be avoided altogether as an alternative. We identified 26 alternative samples with no detectable fragrance chemicals.

Of the 34 conventional samples with detectable fragrance chemicals, 22 contained a product labeled with “fragrance” or other similar descriptors (e.g. “parfum”). Products that contained fragrance chemicals with no label indication were generally cleaners. Of the 17 alternative samples with detectable fragrance chemicals, 14 did not include “fragrance” or similar descriptors as ingredients. The other 3 were labeled “essential oil fragrance” or “plant based fragrance” and contained only natural fragrance compounds. Only one sample, sunscreen, of the 26 alternative samples with no detectable fragrance compounds, had a reference to “fragrance” on the label, specifically listing “fragrance oil blend.”

Glycol ethers

Glycol ethers, a chemical class with more than 80 compounds, are used in a broad array of cleaning applications because of their combined hydrophilic and lipophilic nature. They are often used in paints, varnishes, and cosmetics and have been detected in a variety of household products (Kwon et al. 2008; Plaisance et al. 2008). Biomonitoring methods are currently being

developed, so large-scale studies are limited. Exposure to glycol ethers has been associated in human studies with low sperm mobility (Cherry et al. 2008), hematological effects (Starek et al. 2008), and asthma and allergies (Choi et al. 2010).

In this study, we analyzed all samples for 2-butoxyethanol and 2,2-methoxyethoxyethanol, and in a later second sampling round, we analyzed 14 additional samples for 6 other glycol ethers. We detected glycol ethers in 3 conventional cleaners, face lotion, polish/wax, sunscreen, and in alternative shaving cream, pillow protector, and sunscreen samples. Of the 5 conventional samples with detectable 2-butoxyethanol, only the carpet cleaner was labeled with 2-butoxyethanol. When analyzed and detected, other glycol ethers were not listed on labels. While we detected phenoxyethanol in conventional and alternative sunscreen samples, we did not detect this chemical in some conventional and alternative samples made up of products labeled as containing this compound; possibly levels were below detection limits.

Cyclosiloxanes

Cyclosiloxanes, otherwise known as cyclic volatile methylsiloxanes (cVMSs), are added to consumer products to enhance conditioning and spreading (Silicones Environmental Health and Safety Council of North America 2011). Cyclosiloxanes are widely used, with the most common types being hexamethylcyclotrisiloxane (D3), octamethylcyclotetrasiloxane (D4), decamethylcyclopentasiloxane (D5), and dodecamethylcyclohexylsiloxane (D6). They have been found in cleaning products, personal care products, and baby products at concentrations as high as 1010 µg/g (Horii and Kannan 2008; Wang et al. 2009). Cyclosiloxanes appear to be persistent and have relatively long half-lives in humans. D4 has been shown to have weak estrogenic potential (Quinn et al. 2007) and D5 to be potentially carcinogenic in rats (Wang et al. 2009).

Cyclosiloxanes were analyzed in 10 product types added during the second sampling round. All three cyclosiloxanes were detected in the alternative composite sunscreen, with D5 and D6 at $>4000 \mu\text{g/g}$ and the conventional car interior cleaner at $<100 \mu\text{g/g}$. One cyclosiloxane was detected in the conventional sunscreen (D5; $50 \mu\text{g/g}$) and alternative shaving cream (D6; $10 \mu\text{g/g}$). No product analyzed for cyclosiloxanes indicated “siloxane” on the label; however, two alternative sunscreens were labeled cyclomethicone, a generic name for polydimethylsiloxane, which includes D4, D5 and D6.

UV filters

Organic compounds acting as UV filters are added to many personal care products for skin protection and product stability. Three UV filters in this study, benzophenone-3 (BP-3; oxybenzone), octyl dimethyl PABA, and octinoxate (octyl methoxycinnamate), were detected in a previous study of 75 sunscreen products from European and US manufacturers (Rastogi 2002). Biomonitoring data indicate widespread exposure to some UV filters; BP-3 was detected in 96% of urine samples in the 2003-2004 NHANES (Calafat et al. 2008a). Benzophenone-1 (BP-1), BP-3 and octinoxate are estrogenic *in vitro* and *in vivo* (Schlumpf et al. 2004) and act additively as mixtures (Kunz and Fent 2006).

We analyzed UV filters in sunscreens and the 8 other samples added during the second analytical round. We detected them at $>1\%$ concentration in conventional and alternative sunscreen samples where they were labeled as active ingredients. We detected lower concentrations of three UV filters in conventional sunscreen and shaving cream and in 4 of 5 alternative sunscreens, and none of these were labeled.

Mixtures: Chemicals That Co-Occur Within and Across Products

Our results show that one product can be a source of many chemicals of interest and that use of multiple products can result in exposure to an even larger number of chemicals.

We detected zero to 22 analytes in a single product type (Figure 1). For composited samples, we do not know how many chemicals were in any one of the products; for alternative products, the number of detects ranged up to 11 analytes in shaving cream (Figure 2) and 17 in an individual sunscreen (Figure S1), illustrating the exposure to multiple compounds from a single product.

We identified chemicals that co-occur within a product type by estimating Kendall's τ correlation coefficients (Figure S2). Many fragrance compounds were significantly correlated with each other and with DEP. For example, the natural fragrance limonene was correlated with natural fragrances linalool ($\tau_{\text{conventional}} = 0.43$; $\tau_{\text{alternative}} = 0.59$) and pinene ($\tau_{\text{conventional}} = 0.52$; $\tau_{\text{alternative}} = 0.52$) in both conventional and alternative samples. Limonene and linalool also were positively correlated with DEP in both conventional and alternative samples ($\tau = 0.31$ -0.52). In the conventional samples, DEP was positively correlated with several fragrance compounds (AHTN, benzyl acetate, bucinal, hexyl cinnemal, HHCB, linalool, limonene, and methyl ionone; $\tau = 0.34$ -0.56), which supports the idea that DEP is a common carrier for fragrances. The finding of positive correlations among the fragrance compounds may be influenced by compositing. For example, if each individual product within a product type uses a different fragrance, these compounds will be correlated in the composites, even though an individual product may contain only one of the compounds. We also found that NP and DEHP were correlated in conventional samples ($\tau = 0.4$), consistent with use of both compounds as plastics additives. NP1EO and NP2EO were positively correlated ($\tau_{\text{conventional}} = 0.35$; $p=0.1$), consistent

with their presence in commercial APEO mixtures. In alternative samples, methyl paraben was positively correlated with all three cyclosiloxanes ($\tau = 0.69-0.87$), and the cyclosiloxanes were positively correlated with each other ($\tau = 0.62-0.73$).

Our results also indicate use of multiple products can lead to exposure to an even larger mixture of compounds, even if a consumer selected products considered alternative according to our criteria. For example, a consumer who used the alternative surface cleaner, tub and tile cleaner, laundry detergent, bar soap, shampoo and conditioner, facial cleanser and lotion, and toothpaste (a plausible array of product types for an individual) would potentially be exposed to at least 19 compounds: 2 parabens, 3 phthalates, MEA, DEA, 5 alkylphenols, and 7 fragrances.

The impact of exposures via one product or multiple products is of concern because of the potential combined effects of EDCs or compounds associated with asthma. Our analysis demonstrates that chemical combinations are common in consumer products, and results highlight combinations for toxicity testing, risk assessment, and epidemiological study. Toxicity testing can identify common modes of action for co-occurring chemicals, and risk assessment can then consider cumulative exposures to multiple chemicals. Considering effects of co-occurring compounds in risk assessment would advance the recommendations of the National Research Council (NRC 2008). Similarly, in epidemiological studies, co-occurring exposures need to be understood together, because they may have additive or interacting effects or result in confounding. As an example of possible confounding, several studies have shown an association of endocrine-related health effects with DEP, which does not show activity in animal studies (Duty et al. 2003; Hauser et al. 2007; Lopez-Carrillo et al. 2010; Swan et al. 2005); instead, DEP could be a marker for a large number of synthetic and natural fragrances, which do have activity. This suggests an important area for future research is to characterize the endocrine activity of

fragrances and to measure these compounds in epidemiology studies. Epidemiological studies should include collaborations with toxicologists to help design and interpret findings.

Variability Within Product Types

While our study was not designed to focus on variability in the composition of different individual products within a type, we examined this question for sunscreens. This provides some information about how exposure may differ depending on brand selection. It also allows us to investigate the effect of compositing, which is discussed below as a limitation.

Using sunscreens as an example, we see substantial variability in composition of products within this product type (Figure S1). Among the 5 alternative sunscreens, the number of compounds detected ranged from 4 to 17 per sample, with a total of 24 chemicals detected in the sunscreens. The product with the highest number of detects was marketed for children and favorably rated by a popular environmental health site. The variable composition of individual products within a class is important to consider in exposure modeling and in epidemiologic studies that rely on self-reported product use as a proxy for exposure.

Limitations

To our knowledge, this study is one of the first to look for a large and varied suite of compounds in a broad range of product types; however, the product types and chemicals we included are still a small fraction of those in use, so this report is not comprehensive. In addition, the alternative and conventional products in this study may not be representative. In particular, alternative products, selected according to criteria in Table 1, were mostly purchased at one store with its own criteria, and we do not know how these criteria influenced our product

selection. All products were purchased in 2007-2008 and changing formulations may mean that products purchased today would be different.

We chose to composite conventional products to increase representativeness and limit analytical costs; however, this strategy limits interpretation in several ways. First, compositing does not allow observation of extreme high and low concentrations as it is meant to optimize the estimate of the average concentration. Second, compositing may increase or decrease the number of compounds detected. Twelve chemical concentrations in individual sunscreen samples were diluted to lower concentration categories in the calculated composite, including to values below the DL (Figure S1). Conversely, the number of detects could increase if manufacturers use different chemicals to achieve a particular function in the product, e.g. scent, thereby increasing the number of different chemicals in a composite. To evaluate the effects of compositing on number of detects, we calculated composites from varying numbers of individual sunscreens. The number of detected chemicals in possible sunscreen composites ranged from 5 to 21 (Figure S3) and was positively correlated with the number of products in the composite (Figure S4). These results indicate the varying chemical formulations within a product type. Finally, because we composited conventional and not alternative products and the composites comprise varying numbers of products per sample, direct comparisons between conventional and alternative products and some comparisons between conventional product types could be misleading.

Conclusions

We tested an exceptionally wide range of products, including 50 types of personal care and cleaning products and selected household goods for 66 compounds identified as EDCs or asthma-related. We detected 55 compounds, suggesting a wide range of exposures from common products. Results suggest that vinyl products are an important source of DEHP in

homes. In other products, the highest concentrations and numbers of detects were in the fragranced products perfume, air fresheners, and dryer sheets, and in sunscreen. To our knowledge this is the first test of sunscreens for a range of EDCs. In addition to the labeled ingredients, sunscreens contained up to 7 target chemicals that were unlabeled. The highest number of detects in sunscreen was in a product favorably rated by a popular environmental health website and marketed for babies, children, and sensitive adults, illustrating the limitations of rating products based on ingredients disclosed on product labels. In addition to a broad assessment of chemicals in widely-used personal care and cleaning products, one of our goals was to identify a strategy for reducing exposure by removing or substituting products. Our shopping criteria did identify a set of alternative products with no BPA and antimicrobials and limited synthetic fragrance. We detected DCP, DINP, and DPP only in alternative products, suggesting the possibility that manufacturers have substituted these anti-androgenic phthalates for the better known and also anti-androgenic phthalates DEHP, DBP, and BBP, which are common in conventional products. Our observations of multiple chemicals of concern in composites of high-market-share products coupled with consumers' use of multiple product types—for example, laundry detergent plus dish soap plus shampoo and so on—highlight the importance of considering the cumulative toxicologic effects of combined exposures. Our correlation analysis identifies mixtures for evaluation and also raises cautions that associations in epidemiologic studies may be due to unmeasured chemicals that co-occur with the study target. Disclosure of product ingredients would enable researchers to identify exposures for study and risk evaluation and allow consumers to make decisions consistent with their personal values.

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Table 1. Alternative product selection criteria: List of label terms used to exclude products

Label Term	Reason
parabens	EDC (Kang et al. 2002)
ethanolamines	asthma-related (Kamijo et al. 2009; Makela et al. 2011; Piipari et al. 1998; Savonius et al. 1994)
1,4-dichlorobenzene	carcinogen (IARC 1999)
nonionic surfactants	suggest alkylphenol-based ingredients, which are EDCs (Jie et al. 2010)
fragrances other than “natural fragrances” ^a	asthma-related (Kumar et al. 1995) and EDC (Bitsch et al. 2002; Seinen et al. 1999)
tea tree oil, lavender	EDC (Henley et al. 2007)
triclosan, triclocarban	EDC (Chen et al. 2008; Stoker et al. 2010)
anti-bacterial	suggesting the presence of triclosan or triclocarban
stain-resistant	suggesting organofluorines
vinyl	assumed to contain phthalates
petroleum-based	health concerns about petroleum derivatives

^a“natural fragrances” includes ingredients labeled as essential oils, plant-based fragrances, and other similar ingredients. These were allowed although some individuals may be sensitive.

FIGURE LEGENDS

Figure 1. Concentrations of target compounds in “conventional” consumer products by product type. Compounds are grouped by chemical class, with natural and synthetic fragrances distinguished by dashed horizontal line. Numbers in parentheses after product type indicate number of products in composite. Numbers in the top margin count the number of chemicals detected in each product type; numbers in the right margin count number of products containing each compound. The first 27 product types (left of the solid line) and the last product type are the same in Figures 1 and 2; the remaining product types differ.

Figure 2. Concentrations of target compounds in “alternative” consumer products by product type. Compounds are grouped by chemical class, with natural and synthetic fragrances distinguished by dashed horizontal line. Numbers in the top margin count the number of chemicals detected in each product type; numbers in the right margin count number of products containing each compound. The first 27 product types (left of the solid line) and the last product type are the same in Figures 1 and 2; the remaining product types differ.



